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PHYSICAL AND ELECTRONIC PROPERTIES CHANGED BY AGING PLUTONIUM

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1 INTRODUCTION

Plutonium, because of its radioactive nature, ages from the "inside out" by means of self-irradiation damage and thus produces Frankel-type defects and defect clusters. The defects resulting from the residual lattice damage and helium in-growth could result in microstructural, electronic, and physical property changes. This paper presents volume, density, and electronic property change observed from both naturally and accelerated aged plutonium alloys. Accelerated alloys are plutonium alloys with a fraction of Pu-238 to accelerate the aging process by approximately 18 times the rate of unaged weapons-grade plutonium. After thirty-five equivalent years of aging on accelerated alloys, the samples have swelled in volume by approximately 0.1% and now exhibit a near linear volume increase due to helium in-growth. We will correlate the physical property changes to the electronic structure of plutonium observed by the resonant photoelectron spectroscopy (RESPES).

2 METHOD AND RESULTS

We designed and installed a dilatometry system to monitor long-term growth resulting from the residual lattice damage and helium in-growth in Pu-238 spiked alloys. The dilatometry design is based on linear variable displacement transducer (LVDT) technology that will provide a continuous record of sample length change[1,2]. Figure 1 shows the volume change (ΔV) normalized with the initial volume (V) of spiked alloys at 35°C. The time is represented as an equivalent time (in years) obtained by multiplying the measurement time by the accelerating factor of 18.59. This accelerating factor is obtained by the decay rate of spiked alloy normalized to the reference alloy. This factor will decrease as the material ages due primarily to in-growth of Am-241 in the reference alloy. During the early stage of measurement, the sample's volume increases as a result of self-irradiation and follows the inverse exponential-type of expansion on dose (or time). After the initial expansion, the volume exhibits significantly lower rate of increase and at a near linear expansion behaviour attributed to the helium in-growth mechanism.

Shown in Figure 2 are RESPES results from polycrystalline samples of δ-Pu(Ga)[3]. A significant spectroscopic difference can be observed between the young and aged δ-

Pu(Ga). This result suggests that the valence electronic structure of Pu is dependent upon its microstructure.

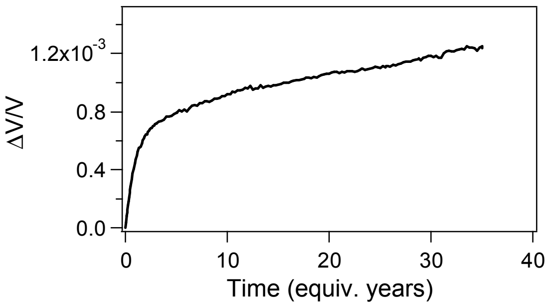

Figure 1. *The volume change normalized with the initial volume of spiked alloy at 35°C.*

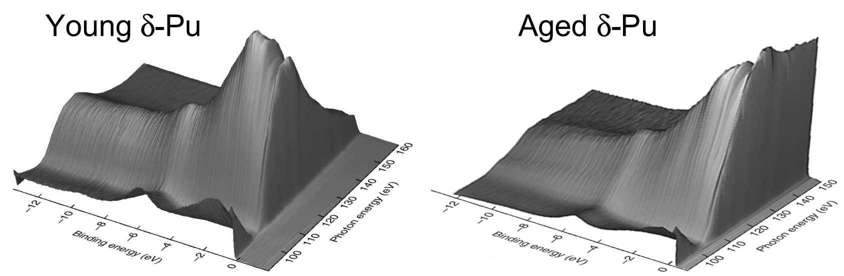

Figure 2. *RESPES comparison between young and aged δ-Pu(Ga)*[3].

3 CONCLUSION

We have shown age-dependent physical and electronic structure of plutonium. Self-irradiation damage causes both lattice damage and helium in-growth that increases volume (or decreases density) in plutonium. RESPES shows differences at the Fermi energy between young and aged δ-Pu(Ga) .

4 ACKNOWLEDGEMENTS

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5 REFERENCES

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